Natural thermoluminescence in some feldspars

A. B. AHMED, R. K. GARTIA, P. S. MAZUMDAR Department of Physics, Manipur University, Imphal 795003, India

Natural thermoluminescence (NTL) in some feldspars namely microcline, orthoclase, albite and oligoclase has been investigated. The study reveals the existence of as many as four peaks in the NTL of both microline and albite in the temperature range 200 to 400° C, whereas in the case of orthoclase three peaks could be located. The results are discussed in the light of the findings of the previous workers. Some of the NTL peaks observed in this study are analysed by curve fitting method, and their trapping parameters, namely thermal activation energy (E), frequency factor ($s'n_0$) and order of kinetics (b) established. A one to one correspondence between these peaks and the ones observed in the laboratory X-ray-irradiated sample is demonstrated.

1. Introduction

The feldspars form an important group of rock forming silicates and most of the members exhibit good natural thermoluminescence (NTL). Some of these materials have been already found to be suitable candidates for application in geological problems such as dating and state of weathering of balsalts [1]. The use of feldspars in TL dating has been done by Gillot et al. [2]. May [3] has recommended feldspars for dating basalts. Ogelman and Kapur [4], who studied the TL in feldspars from basalts undergoing weathering, have noticed that with the increasing presence of kaolinite, a clay mineral derived from alteration of feldspars during weathering, the TL efficiency also increases. This feature could be useful in assessing the state of weathering. Angino [5] reported the effect of shearing pressure on the NTL of several minerals including orthoclase, and suggested that detailed TL studies of pressure-induced changes in the glow curve characteristics of different rock and mineral species may be a possible tool for investigating pressure and temperature distribution both in areas of intrusive rocks and rock materials surrounding an underground nuclear explosion. Hence study on NTL of the feldspars is of practical interest.

NTL in feldspars have been reported by a number of investigators and most of them observed peak in the 200 to 400° C region – a brief summary of which can be found in the article of McDougall [6]. In a subsequent paper Nishita *et al.* [7] have reported NTL of some feldspars. In spite of these studies [6, 7], the existing knowledge on the NTL of feldspar remains still imperfect so far as the number of NTL peaks, the peak temperature and their characteristic parameters are concerned. In this paper we have attempted to depict a comprehensive picture of NTL observed in some feldspars in the light of the previous studies on these materials [6, 7] including those of X-rayirradiated TL [8–10].

In our recent study on X-ray-irradiated TL of some feldspars, we have established the existence of a

0022-2461/88 \$03.00 + .12 © 1988 Chapman and Hall Ltd.

number of peaks in the temperature range 200 to 400° C in microline and orthoclase [8], albite [9], and oligoclase [10]. Since qualitatively the NTL and the laboratory X-ray-irradiated TL are expected to be identical, we have tried to establish a correlation between these two in this study.

2. Experimental details

The feldspars used in this study are microcline, orthoclase, albite and oligoclase. Microcline (triclinic form of KAlSi₃O₈) of four different shades include bluish green, green, brown and colourless varieties. The orthoclase (monoclinic KAlSi₃O₈) is of yellowish variety; the two varieties of albite (NaAlSi₃O₈) are one semi-transparent and other opaque. The oligoclase sample used here is white, massive. All the samples are collected from different regions of India, except orthoclase which is of Norwegian origin. Thin pieces (~1 mm) of the samples are separated out of the bulk material without applying much stress. The samples are then washed with alcohol and dried. Natural TL of these samples are recorded using a rate of heating of 0.67 deg sec⁻¹.

3. Results and discussions

The NTL curves of some feldspars are shown in Figs 1, 2a and 3. Fig. 1 shows the NTL pattern of various shades of microcline. Whereas the bluish green variety exhibits three distinct peaks at 190, 300 and around 360°C (curve a, Fig. 1) the green sample gives a single peak at 230° C with unusual broadness (Curve b, Fig. 1). In the brown sample, the prominent peak appears at 280° C preceded by a shoulder around 230°C (Curve c, Fig. 1). The colourless variety also shows the strongest peak at 280°C flanked by two more peaks on either side of 280° C around 200 and 320°C in the form of shoulders (Curve d, Fig. 1). Fig. 2a shows the NTL of orthoclase. The peak of maximum intensity appears at 215°C followed by an ill-defined peak around 280° C. There is indication of a faint peak around 350°C as well. The NTL of two



Figure 1 NTL curves of various shades of microcline, (a) bluish green, (b) green, (c) brown and (d) colourless varieties.

varieties of albite along with that of oligoclase are shown in Fig. 3. The semi-transparent sample exhibits three peaks at 210° C, followed by the dominant one at 310° C and at around 400° C (Curve a, Fig. 3). In the opaque variety, on the other hand, the main peak appears at 240° C with another distinct peak at 310° C

(Curve b, Fig. 3). The complex glow curve of oligoclase has its main peak at 340° C. There is indication of the presence of another around 270° C (Curve c, Fig. 3). It is to be noted that though the NTL curve of green microcline (Curve b, Fig. 1) appears to be elementary peaking at 230° C, the unusual broadness



Figure 2 (a) NTL curve of orthoclase. (b) X-ray-irradiated TL curves of the same obtained by thermally cleaning the low temperature peaks up to 170° C. (—) Experimental curve, (O) numerically computed peak, (–) difference curve.



Figure 3 NTL curves of albite and oligoclase. (a) Semi-transparent and (b) opaque varieties of albite, (c) oligoclase.

however suggests the multipeak nature of the curve. We have established, in a study, that such a glow curve in fact is the result of superposition of several peaks [10]. It is evident from the observation of Figs 1 and 3 that for the same mineral of different varieties, the most intense peak appears at different temperatures. This can be attributed to the fact that the place of their origin are different and hence the defects (associated with impurities, thermal history, pressure, etc.) present in them are expected to vary. TL is an extremely structure-sensitive phenomenon and hence the background impurities in the phosphor is a critical factor in the TL emission. Also, the TL properties exhibited by a phosphor very much depend upon the kind of "thermal annealing" experienced by it prior to the irradiation/excitation. These two aspects, namely the effects of impurities and thermal treatments on TL are discussed by Nambi [11]. Pressure is also a factor which may affect TL [5].

The position of the peaks observed in the NTL of these feldspars along with those of McDougall [6] and Nishita *et al.* [7] are displayed in Table I. Incompatible with the observation of McDougall [6] who reports a single peak at 380° C both in microcline and albite, we observe more than one peak in these systems. In their study, Nishita *et al.* [7] report the multi-peak structure in the glow curve of feldspars which is consistent with our result. However, in the present investigation, a few additional peaks are observed which are not reported by these authors. In this context, it may be mentioned that in our previous studies on X-ray-irradiated TL of these feldspars, we have already demonstrated the existence of a number of peaks in the temperature range 200 to 400° C [8–10]. All the peaks observed in the NTL glow curve of the samples have also been found to appear in the X-ray-irradiated TL more or less in same temperature region. As for example, in the NTL of orthoclase two peaks are observed at 215 and around 285°C with the indication of the presence of another weak one around 350°C (Fig. 2). In the X-ray-irradiated TL the existence of peaks around 202, 280 and 350°C in the system is already established [8].

A look into the Table I reveals that there is inconsistency in the peak temperature of the present ones with those of the earlier researchers [6, 7]. Such a thing is not unusual since the glow curve results depend on heating rate both qualitatively and quantitatively which is discussed in detail by Chen and Kirsh [12]. The previous authors [6, 7], most probably because of this, could not observe all the peaks as the heating rates in their studies were rather high (Table I). Angino [5] has studied the effects of shearing pressure on the NTL of several minerals. He reports that the

TABLE I Natural TL peaks in some feldspars

Sample	McDougall [6] Peak temperature (° C) $(\beta = 21.10^{\circ} \text{ C sec}^{-1})$	Nishita et al. [7]	Present study		
		Peak temperature (° C) ($\beta = 8.33^{\circ} \text{ C sec}^{-1}$)	Variety	Peak temperature (° C) ($\beta = 0.67^{\circ} \text{ C sec}^{-1}$)	
microcline			bluish-green	190*, 300, 360	
	380	283, 370	green	230	
			brown	230, 280*	
			colourless	200, 280*, 320	
orthoclase			yellowish	215*, 280, 350	
albite			semi-transparent	210, 310*, 400	
	380	320, 400	opaque	240*, 310, 366 [†]	
oligoclase		300, 425	white	270, 340*	

* Most intense peak.

[†]Peak observed after analysis of the previous peaks.



Figure 4 Total curve fitting of the theoretical curve to the experimental one with three secondorder peaks in albite. (a) NTL curve, (b) X-rayirradiated TL obtained by thermally cleaning the low temperature peaks up to 220° C. (-----) Experimental curve, (O) numerically computed peak, (\bullet) total synthesized curve.

only peak occuring at 255° C in orthoclase, with the application of pressure, initially shifts upward, thereafter with the increase of pressure, the peak temperature shifts slowly downward through the intensity of the peak remains essentially unchanged.

The fact that both NTL and X-ray-irradiated TL peaks appear in the same temperature range leads to the inference that there exists one to one correspondence between these two sets of peaks. In an attempt to establish a correlation between these two, some of the major NTL peaks are analysed by curve fitting method using the following second order kinet-

ics equation of Garlick and Gibson [13]:

$$I = s' n_0^2 \exp\left(-\frac{E}{kT}\right) \\ \times \left[1 + \frac{s' n_0}{\beta} \int_{T_0}^T \exp\left(-\frac{E}{kT}\right) dT\right]^{-2}$$
(1)

and the condition of maximum is:

$$1 + \frac{s'n_0}{\beta} \int_{T_0}^{T_m} \exp\left(-\frac{E}{kT}\right) dT$$
$$= \frac{2kT_m^2 s'n_0}{\beta E} \exp\left(-\frac{E}{kT_m}\right)$$
(2)

TABLE II Trapping parameters of some TL peaks in some feldspars

Sample	NTL			X-ray-irradiated TL		
	<i>T</i> _m (° C)	E (eV)	$\frac{s'n_0}{(\sec^{-1})}$	$\frac{T_{\rm m}}{(^{\rm o}{\rm C})}$	E (eV)	$s'n_0$ (sec ⁻¹)
Bluish-green microcline	190	1.06	1.25×10^{10}	186	1.08	2.78×10^{10}
Brown microcline	280	1.36	8.10×10^{10}	275	1.36	1.05×10^{11}
Yellowish orthoclase	215	1.12	1.34×10^{10}	202	1.16	7.68×10^{10}
Opaque albite	240	1.26	8.90×10^{10}	250	1.24	1.55×10^{10}
	310	1.48	1.99×10^{12}	307	1.36	1.68×10^{10}
	366	1.56	4.60×10^{10}	352	1.52	4.60×10^{10}

where I is the TL intensity, $s'n_0$ is the frequency factor (s' being the pre-exponential factor, a constant with dimension cm³ sec⁻¹) and n_0 the initial concentration of trapped charge carriers (cm^{-3}) E the thermal activation energy of trap depth (eV), T_0 the initial temperature (K), $T_{\rm m}$ the peak temperature (K) and k the Boltzmann constant (eVK^{-1}). In order to determine the trapping parameters namely E and $s'n_0$ the theoretical peaks are numerically generated using the condition of maximum and the parameters thus extracted are set in Table II along the corresponding values for X-ray-irradiated samples. In the computation of the theoretical peaks we have assumed second-order kinetics process (i.e. order of kinetics b = 2) since all the X-ray-irradiated TL peaks in these systems are known to obey second-order kinetics [8-10]. The fitting of the computed peaks with those of the experimental ones is good and some of these are shown in Figs 2a and 4a for orthoclase and albite, respectively. The corresponding fitting of the X-ray-irradiated peaks are also depicted in Figs 2b and 4b. Though the relative intensity of the NTL and TL peaks are different, a comparison of the trapping parameters of the NTL peaks to those of the corresponding peaks of the X-ray-irradiated samples (Table II) shows good agreement in their values which clearly demonstrates the one to one correspondence between the two sets of peak.

The present investigation on the NTL of some feldspars demonstrates the following.

1. Contrary to the observation of McDougall [6] who reports a single peak in the NTL of microcline and albite, as many as four peaks could be located both in microcline, and albite in the temperature range 200 to 400° C. The multipeak structure of the NTL pattern as reported by Nishita *et al.* [7] agrees with the present findings. However, the present study has

revealed the existence of peaks in addition to those reported by these authors [6, 7].

2. Since in general the NTL of feldspars consists of more than one peak it is suggested that while utilizing their NTL in any geological/archaeological applications like dating, one has to consider the effect of the satellite peaks, because in some cases their contribution may lead to erroneous results.

3. There is a one to one correspondence between the NTL and laboratory X-ray-irradiated TL peaks.

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Received 4 December 1986 and accepted 15 May 1987